

DISCUSSION OF THE AMENDMENT

Due to the length of the specification herein, Applicants will cite to the paragraph number of the published patent application (PG Pub) of the present application, i.e., US 2007/0225462, when discussing the application description, rather than to page and line of the specification as filed.

Claim 1 has been amended by incorporating the subject matter of Claim 5 therein, and further amended to exclude, in effect, non-cyclic monoethers from the definition of General formula [1]. Claim 5 has been canceled.

Claim 15 has been amended by incorporating that part of the subject matter of Claim 19 limited to ketones therein. Claim 19 has been canceled.

New Claim 23 has been added, dependent on Claim 15 and limiting the ketone to methyl ethyl ketone or methyl isobutyl ketone, as supported in the specification at paragraph [0074].

No new matter is believed to have been added by the above amendment. Claims 1-4, 6-18 and 20-23 are now pending in the application.

REMARKS

The rejections under 35 U.S.C. § 103(a) of:

Claims 1-12 and 15-22 as unpatentable over JP 2003-277308 (Honda) in view of US 3,814,713 (Honda et al) and JP 57-143316 (JP Okamura et al);

Claim 13 as unpatentable over Honda in view of Honda et al and Okamura et al, and further in view of US 5,368,928 (US Okamura et al); and

Claim 14 as unpatentable over Honda in view of Honda et al and Okamura et al, and further in view of US 2004/0116647 (Swedo),

are respectfully traversed.

As recited in above-Claim 1, an embodiment of the present invention is a process for producing a resorcinol-formalin resin containing no inorganic salts, which comprises:

(1) adding, to a water solvent, resorcinol (A) in an amount of 20 to 150 parts by weight relative to 100 parts by weight of water, an inorganic salt (B) in an amount of 20 to 80 parts by weight relative to 100 parts by weight of water, and an organic solvent (C) having a solubility parameter of 7.0 to 12.5 and capable of dissolving the resorcinol-formalin resin in an amount of 10 to 200 parts by weight relative to 100 parts by weight of resorcinol (A), thereby forming a mixture;

(2) stirring the mixture at a liquid temperature not higher than the boiling point of the organic solvent (C) to give a two-phase system containing no remaining solid matter;

(3) adding a catalytic amount of an organic acid or inorganic acid (D) to the mixture formed in step (2);

(4) adding 1 to 40% formalin (E) dropwise to the mixture formed in step (3) in a molar ratio of formaldehyde/resorcinol of 0.3 to 0.8 under stirring over a period of 1 to 300 minutes thereby forming a reaction system, while maintaining the reaction system at 0 to 60°C;

(5) stirring the mixture formed in step (4) for further 10 to 60 minutes after the completion of the dropwise addition to cause a liquid-liquid heterogeneous reaction to proceed;

(6) allowing the reaction system to stand while maintaining it at the temperature of the reaction to separate it into two layers, which are an aqueous layer and a reaction product layer;

(7) removing the aqueous layer;

(8) adding an organic solvent (C) in an amount of 1 to 5 equivalents to the amount of the reaction product to the reaction product layer which is an organic solvent layer to effect dilution;

(9) adding water to the reaction product layer in an amount which is half of the amount of the organic solvent;

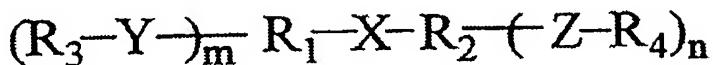
(10) stirring the reaction system after adding of said organic solvent (C) and said water while maintaining its temperature to be not higher than the boiling point;

(11) separating the reaction system of step (10) into two layers, including an aqueous layer, after allowing the reaction system to stand; and then

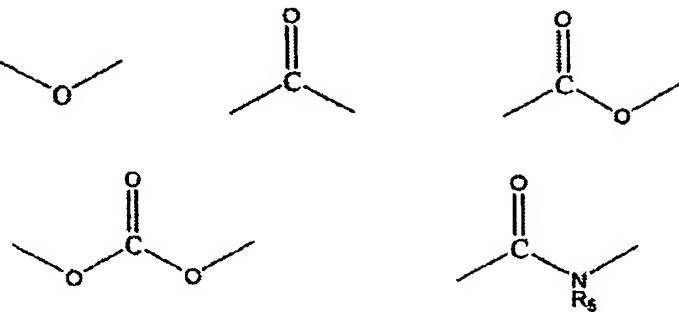
(12) removing the aqueous layer to obtain a resorcinol-formalin resin,  
wherein said reaction is a one-stage reaction and liquid-liquid distribution is conducted in the same reactor, and

wherein the organic solvent (C) is an organic solvent represented by the following general formula [1]:

General formula [1]:



X , Y , Z :



wherein m represents 0 or 1, n represents 0 or 1, and R<sub>1</sub>, R<sub>2</sub>, R<sub>3</sub>, R<sub>4</sub>, and R<sub>5</sub> each independently represents a methyl group, an ethyl group, an n-propyl group, a 2-propyl group, an n-butyl group, a secondary butyl group, an isobutyl group, or a tertiary butyl group; R<sub>1</sub> and R<sub>2</sub> may be combined to form a ring when m=n=0, R<sub>2</sub> and R<sub>3</sub> may be combined to form a ring when m=1 and n=0, and R<sub>3</sub> and R<sub>4</sub> may be combined to form a ring when m=n=1, with the proviso that when X is -O- and m=n=0, R<sub>1</sub> and R<sub>2</sub> are combined to form a ring.

Another embodiment of the present invention, as recited in Claim 15, is, in effect, the product made by the process of Claim 1, except that the organic solvent (C) is limited to a ketone.

In sum, an embodiment of the present invention is directed to a process comprising: adding, to a water solvent, resorcinol, an inorganic salt (molecular weight controlling agent), and a specific organic solvent (C) (having a solubility parameter of 7.0 to 12.5); stirring the resulting mixture to give a two-phase system; adding an acid catalyst; dropwise adding formalin thereto; stirring the resulting mixture to cause a liquid-liquid heterogeneous reaction. The process of the invention provides a resorcinol-formalin resin containing no inorganic salts, having a moderate flowability, and having reduced contents of resorcinol monomer and resorcinol pentanuclear or higher nuclear bodies. Another embodiment of the

present invention is directed to a product made by the process, except that the organic solvent (C) is limited to a ketone. As discussed in further detail later, the product claims are separately patentable.

As described in the specification under Background Art, beginning at paragraph [0002], resorcinol-formalin resins are known in the art, but flowability has been problematical, as well as the suggested solutions thereof. An object of the present invention, as described in the specification at paragraph [0006], is to provide a process for producing a resorcinol-formalin resin containing no salts and having a moderate flowability when transformed into an aqueous solution, which is capable of reducing both the content of resorcinol monomer and the content of resorcinol-formalin resin of resorcinol pentanuclear or higher nuclear bodies in a one-stage reaction. Applicants have been able to attain this objective by adopting a liquid-liquid heterogeneous reaction using a specific range of organic solvents, i.e., organic solvents having a solubility parameter within a particular range, as described in the specification at paragraph [0007].

A so-called liquid-liquid heterogeneous reaction is one in which the reaction is carried out in a mixed solvent of water and a solvent. In the present invention, it is essential to use the organic solvent.

Honda discloses a low-molecular weight resorcinol-formaldehyde reaction product having a small content of unreacted resorcinol and excellent flowability, wherein resorcinol and formaldehyde are reacted in the presence of an acidic catalyst, a molecular weight controlling agent [0009] is added and the reaction is carried out in water. The kind and amount of the molecular weight controlling agent, which may be a particular organic or inorganic salt [0013], are selected so that a large amount of dimer and trimer of resorcinol is precipitated. A precipitated product generated when the concentration of the generated

substance reaches the saturated concentration and no further dissolution in water is possible is collected.

For collecting the reaction product, it is good to squeeze out the mother liquid, for example, by squeezing under pressure, but this cannot completely remove the mother liquid. If necessary, the product is washed with a resorcinol washing liquid. Further, if necessary, a selected solvent method using a solvent that dissolves the reaction product but does not dissolve the molecular weight controlling agent is used. When the molecular weight controlling agent is calcium chloride, 2-butanone may be used for the selected solvent method [0021] and [0022].

Thus, according to Honda, the reaction of resorcinol and formaldehyde is carried out in an **aqueous solvent** in the presence of a molecular weight controlling agent, such as calcium chloride. The low-molecular weight resorcinol-formaldehyde reaction product is obtained as a precipitate generated when the concentration reaches the saturated concentration and no further dissolution becomes possible. In the present invention, on the other hand, a specific organic solvent having a solubility parameter falling within a specific range and immiscible with water, and a heterogeneous system reaction is carried out in the presence of an inorganic salt, such as calcium chloride, serving as a molecular weight controlling agent.

According to Honda, the reaction product precipitates after reaching the saturated concentration, because no further dissolution in water is possible. In the present invention, on the other hand, since an organic solvent, such as methyl ethyl ketone (MEK), having dissolution ability with respect to the reaction product as compared with water is used, precipitation does not occur and the reaction product migrates to the organic solvent side.

The resorcinol-formalin resin obtained by the present invention has a balanced distribution over the whole molecular weight region from low molecular weight (resorcinol

monomer) to high molecular weight (pentanuclear or higher nuclear bodies) as compared to the resin obtained by Honda, and further, has flowability. Furthermore, the objective low-molecular weight resorcinol-formalin resin is dissolved in the organic solvent, and does not contain an inorganic salt such as calcium chloride serving as the molecular weight controlling agent. For this reason, the product shows an excellent effect in adhesion with another material.

Moreover, the process of the invention does not require, as Honda requires, “squeezing under pressure”, “washing with a resorcinol washing liquid” or “selected solvent method” in collecting the reaction product.

Honda et al discloses an adhesive composition for bonding rubber materials to any of various materials including synthetic and natural polymers, metals, silicate and ceramic materials with only a single application, which composition comprises a rubber latex, a high molecular weight resorcinol-formaldehyde resin and a low molecular weight resorcinol-formaldehyde resin, wherein the latter component contains unreacted resorcinol in an amount less than 15 wt% (Abstract). The low molecular weight resorcinol-formaldehyde (RF) resin is disclosed as “must be prepared carefully according to a particular process” (sentence bridging columns 2 and 3). This component is obtained by reacting resorcinol with formaldehyde in a mole ratio of 1 to 0.05-0.8, preferably in water and without a catalyst or in the presence of a very small amount of an acidic catalyst (column 4, lines 6-12). Honda et al discloses further that in order to synthesize the low molecular weight RF resin involving very little or no amount of resorcinol polymers higher than the pentamer inclusive, it is necessary to react resorcinol with formaldehyde while limiting the amount of the latter to less than equimole of the former (column 4, lines 34-38). After completion of the reaction, the reaction product is dissolved in a ketone, and the extract is repeatedly washed with water or extracted therewith so that unreacted resorcinol is selectively transferred to the water phase,

thereby resulting in RF resin having unreacted resorcinol in an amount decreased down to a predetermined ratio, wherein methyl isobutyl ketone and diisobutyl ketone are the preferred ketones, which ketones are used in an amount of 4 to 6 times the lower molecular weight RF resin (column 5, lines 1-43).

Thus, Honda et al uses the ketone solvent in a post-treatment after completion of the reaction. By the repeated water extractions from the organic solvent layer in which the reaction product is dissolved, the content of the unreacted resorcinol remaining in the resorcinol-formaldehyde resin is reduced to less than 15 wt%.

As discussed above, the ketone solvent is used for the post-treatment after completion of the reaction. The reaction *per se* is carried out in an **aqueous system**. The reaction system is also different from that of the present invention. Further, the low-molecular weight resorcinol-formaldehyde resin and the high-molecular weight resorcinol-formaldehyde resin are prepared under separate conditions, and they are blended at a predetermined ratio to obtain an objective adhesive composition. The present invention, on the other hand, is directed to a production process in which all the steps including the one-stage reaction and the liquid-liquid distribution are carried out in the same single reactor. This provides a further distinction over Honda et al.

In addition, according to the process of the present invention, when using a ketone such as MEK as the organic solvent, the resin and the solvent react to form a structure in which the solvent is incorporated as part of the resin structure, hence providing a ketone-modified resorcinol-formalin resin (novel resin). For this reason, it provides a modified structure that is promising to dramatically increase the adhesion property as compared to conventional resorcinol-formalin resins. This contributes improvements in quality and performance.

JP Okamura et al is directed to a process for producing a resorcinol-formaldehyde resin through a condensation reaction of resorcinol and a formaldehyde compound, in which a solvent is added that dissolves resorcinol but does not substantially dissolve components obtained by the condensation reaction and having a molecular weight of a dinuclear body or higher, to the system either during, before or after the condensation reaction in the presence or absence of water in order for treating the condensate, and then the resin layer is separated from the solvent layer. The solvent is at least one member selected from aromatic hydrocarbons, halogenated hydrocarbons and ethers. Such solvents are not within the terms of presently-recited organic solvent (C).

US Okamura et al is directed to a water-based treating liquid for a rubber-reinforcing glass fiber cord, which contains as a solid component a composition consisting essentially of:

(1) a rubber latex

(2) an alkylated chlorosulfonated polyethylene latex, and

(3) a water-soluble resorcinol-formaldehyde condensate, wherein, based on the total amount of the components (1) and (2), the amount of the component (2) is 10 to 70% by weight and the amount of the component (3) is 5 to 20% by weight (column 3, lines 5-18).

The resorcinol-formaldehyde resin disclosed in US Okamura et al is necessarily a resole-type resin obtained by using an alkali as a condensation agent, which is clearly distinct from a novolak-type resin obtained by using an acidic catalyst, such as hydrochloric acid, as a condensation agent. Also, the production process is different from that of the present invention. US Okamura et al is, in essence, irrelevant.

Swedo relates to a particular phenol resin, and discloses various compounds to be used as a hardener and/or a catalyst for curing a phenol resin. Example 13 therein is drawn to a process for synthesizing a phenol-formalin resin (resole-type resin) in which a “sodium hydroxide solution” is used as a condensation agent. After the reaction in the aqueous

system, hydrochloric acid is added for neutralization, and then the upper, aqueous layer is decanted to obtain a resole layer. The resole is diluted with acetone, and the solution is dried over anhydrous magnesium sulfate, followed by filtration and removal of the acetone, to thereby obtain the resole-type resin. Thus, like US Okamura et al, Swedo is, in essence, irrelevant.

Claim 15 and claims dependent thereon are separately patentable. A feature thereof resides in modification of the resorcinol-formalin resin by the ketone compound solvent. When a ketone solvent is used, the resorcinol-formalin resin is modified with the ketone. The ketone modification contributes significant enhancement of performance such as adhesion between a rubber tire and a polyester resin, as compared to the case of conventional resorcinol-formalin resins. The above is supported in the specification by various Examples. In Examples 1 to 5, using a ketone compound as an organic solvent, calculated yields are from 102 to 111% as summarized in the Table below. A yield over 100% is evidence of the modification with the ketone compound.

On the other hand, tetrahydrofuran (THF) used in Example 6 has an ether structure and does not react with the resorcinol-formalin resin. Further, resins dissolved in tetrahydrofuran partly transfers to the aqueous layer. Hence, the yield is low.

Regarding Examples 7 to 11, the yields cannot be calculated because the amounts of the resins obtained are not disclosed.

Example No.	Solvent	Yield (%)	Remarks
1	MEK	110.7	
2	MEK	107.0	
3	MEK	106.5	
4	MEK	107.0	
5	MIBK	101.7	
6	THF	95.0	Low yield because of miscibility with water

Calculation of Yield (Example 1):

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Yield(%) = (Amount of Resin Solution) × (solid content) × 100/ ((Amount of Resorcinol) + (Amount of Formalin))

$$= 263 \times 0.50 \times 100/(111 + 7.84) = 110.7 (\%)$$

wherein the "Amount of Formalin" =

$$((\text{Charged Amount}) \times (\text{solid content (37\%)})/30 \times \text{dehydration (30-18)}) = (53 \times 0.37)/30 \times 12 = 7.84$$

\*Molecular weight of formalin: 30

\*Molecular weight of water: 18

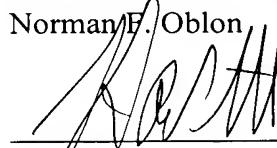
For all the above reasons, it is respectfully requested that the rejections be withdrawn.

All of the presently-pending claims in this application are now believed to be in immediate condition for allowance. Accordingly, the Examiner is respectfully requested to pass this application to issue.

Respectfully submitted,

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